

# Quantum criticality in $A\text{Fe}_2\text{As}_2$ ( $A = \text{K}, \text{Rb}, \text{Cs}$ ) superconductors probed by $^{75}\text{As}$ NMR spectroscopy

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We report  $^{75}\text{As}$  nuclear magnetic resonance measurements on single crystals of  $\text{RbFe}_2\text{As}_2$  and  $\text{CsFe}_2\text{As}_2$ . Taking previously reported results for  $\text{KFe}_2\text{As}_2$  into account, we find that the anisotropic electronic correlations evolve towards a magnetic instability in the  $A\text{Fe}_2\text{As}_2$  series (with  $A = \text{K}, \text{Rb}, \text{Cs}$ ). Upon isovalent substitution with larger alkali ions, a drastic enhancement of the anisotropic nuclear spin-lattice relaxation rate and decreasing Knight shift reveal the formation of pronounced spin fluctuations with stripe-type modulation. Furthermore, a decreasing power-law exponent of the nuclear spin-lattice relaxation rate  $(1/T_1)_{H\parallel ab}$ , probing the in-plane spin fluctuations, evidences an emergent deviation from Fermi-liquid behavior. All these findings clearly indicate that the expansion of the lattice in the  $A\text{Fe}_2\text{As}_2$  series tunes the electronic correlations towards a quantum critical point at the transition to a yet unobserved, magnetically ordered phase.

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The investigation of quantum critical points (QCPs) in iron pnictides provides a route for understanding the interplay of the high superconducting transition temperatures,  $T_c$ , and anomalous normal-state properties, such as enhanced quasiparticle masses and non-Fermi-liquid behavior [1–11]. A magnetic QCP, emerging from the interplay of electronic localization and itinerancy, was theoretically anticipated [11], and has been experimentally identified in  $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$  by various techniques [1–5]. Besides applying chemically induced pressure by P substitution, carrier doping is discussed as another control parameter to drive a QCP in the  $\text{BaFe}_2\text{As}_2$  family.

In the heavily hole-doped materials  $A\text{Fe}_2\text{As}_2$  ( $A = \text{K}, \text{Rb}, \text{Cs}$ ), it is proposed that a QCP is approached with increasing negative pressure by an isovalent replacement of the  $\text{K}^+$  ion with larger  $\text{Rb}^+$  or  $\text{Cs}^+$  [8–10]. In  $\text{KFe}_2\text{As}_2$ , the  $3d$  orbitals are nominally filled with 5.5 electrons, showing a pronounced mass enhancement due to strong correlation effects, as well as deviations from standard Fermi-liquid behavior [12–16]. The increased negative pressure by  $\text{Rb}^+$  and  $\text{Cs}^+$  substitution does not affect the topology of the Fermi surfaces, but leads to a strong increase of quasiparticle masses, probably driven by an orbital-selective increase of correlations [10, 16–20]. In line with these findings, the application of positive pressures was reported to decrease the Sommerfeld coefficient  $\gamma$  in  $\text{KFe}_2\text{As}_2$  [21].

In a recent dilatometry and local-density approximation (LDA) study of the  $A\text{Fe}_2\text{As}_2$  systems, Eilers et al. suggested that the enhancement of correlations originates from a change of the hybridization of Fe  $d_{xy}$  orbitals [10]. Further, the critical fluctuations close to the QCP, possibly

associated with magnetic order related to a Mott-insulating phase, appear to suppress  $T_c$ , in contrast to the often propagated picture of superconductivity stimulated by fluctuations in the vicinity of a QCP, encountered in, e.g., P-doped  $\text{BaFe}_2\text{As}_2$  [5]. In general, approaching a magnetic QCP is reflected by pronounced non-Fermi-liquid behavior and a drastic increase of slowly fluctuating magnetic degrees of freedom, yielding a power-law behavior of related physical quantities with exponents differing from those of a Fermi liquid [22].

The findings of Eilers et al. clearly indicate that a quantum critical point is approached with increasing FeAs-cell volume. However, other experimental reports seem much less conclusive. Different power-law dependencies between  $\propto T^{1.5}$  and  $\propto T^{2.0}$  were reported for the resistivity of the  $A\text{Fe}_2\text{As}_2$  series [12, 16, 17, 23, 24]. An investigation of possible anisotropies of the electrical transport properties might shed light on these seemingly conflicting results, but was not reported so far. From nuclear magnetic resonance (NMR) measurements of all three  $A\text{Fe}_2\text{As}_2$  compounds with  $H \parallel c$ , the same power law of  $1/T_1 \propto T^{0.75}$  was reported, i.e., giving no indication of approaching quantum criticality [25]. In order to resolve this discrepancy between the different experimental findings, we performed a detailed NMR study of the anisotropic magnetic correlations.

In this Letter, we present  $^{75}\text{As}$  NMR measurements on high-quality single crystals of  $\text{RbFe}_2\text{As}_2$  and  $\text{CsFe}_2\text{As}_2$  for both in-plane ( $H \parallel ab$ ) and out-of-plane ( $H \parallel c$ ) orientations of the magnetic field and temperatures between 1.6 and 300 K. Taking previously reported results for  $\text{KFe}_2\text{As}_2$  into account, we find a continuous reduction of

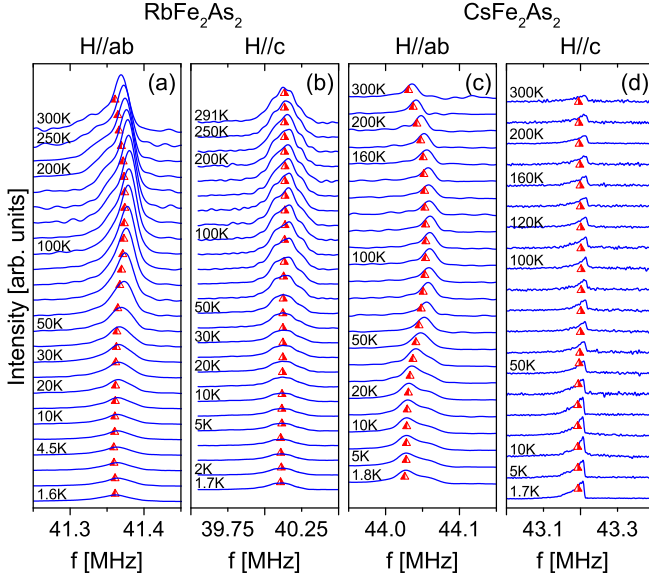


FIG. 1. (a) - (d): The  $^{75}\text{As}$  central transition spectra for  $\text{RbFe}_2\text{As}_2$  and  $\text{CsFe}_2\text{As}_2$  with the magnetic field applied in-plane ( $H \parallel ab$ ) and out-of-plane ( $H \parallel c$ ). Red triangles mark the first spectral moment.

the Knight shift and a drastic increase of spin fluctuations along the  $\text{AFe}_2\text{As}_2$  series. Based on the anisotropy of  $1/T_1$ , we provide evidence for a stripe-type modulation of the spin fluctuations in all three  $\text{AFe}_2\text{As}_2$  compounds. Furthermore, a successively reduced power-law exponent of  $(1/T_1)_{H \parallel ab} \propto T^\eta$  at low temperatures shows increasing deviation from the unity exponent of a Fermi liquid. Our results clearly indicate that the electronic correlations are evolving from proximity to a Fermi liquid towards a state with strong correlations in close vicinity to a QCP.

Single crystals of  $\text{AFe}_2\text{As}_2$  ( $A = \text{Rb}, \text{Cs}$ ) were grown by use of the self-flux technique [24]. Their stoichiometry was confirmed by energy-dispersive x-ray spectroscopy. For a precise sample orientation with field, a single-axis goniometer with a resolution of  $0.1^\circ$  was used. The  $^{75}\text{As}$  NMR central transition spectra and nuclear spin-lattice relaxation rate  $1/T_1$  were measured at temperatures from 1.6 to 300 K at  $\mu_0 H = 5.513$  T for  $\text{RbFe}_2\text{As}_2$  and at  $\mu_0 H = 5.900$  T for  $\text{CsFe}_2\text{As}_2$ .  $T_1$  was obtained from fitting  $M_z(t) = M_0 [1 - f(0.9e^{-6t/T_1} + 0.1e^{-t/T_1})]$  to the recovery of the nuclear magnetization after inversion [26].

By measuring the central ( $I_z = -1/2 \rightarrow +1/2$ ) and satellite ( $-3/2 \rightarrow -1/2, +1/2 \rightarrow +3/2$ ) transition frequencies for  $H \parallel ab$  and  $H \parallel c$ , we determined the principal axis of the local electric field gradient (EFG) tensor  $V_{ZZ}$  to be parallel to the  $c$  axis, and the quadrupole frequency  $\nu_Q \propto V_{ZZ}$  at 5 K as 14.4 MHz for  $\text{RbFe}_2\text{As}_2$  and 13.7 MHz for  $\text{CsFe}_2\text{As}_2$ , larger than  $\nu_Q = 12.2$  MHz, reported for  $\text{KFe}_2\text{As}_2$  [9]. Between 5 to 300 K,  $\nu_Q$  shows, in good accordance with the thermal expansion of the lattice, a weak variation ( $\lesssim 1\%$ ) for both  $\text{RbFe}_2\text{As}_2$  and

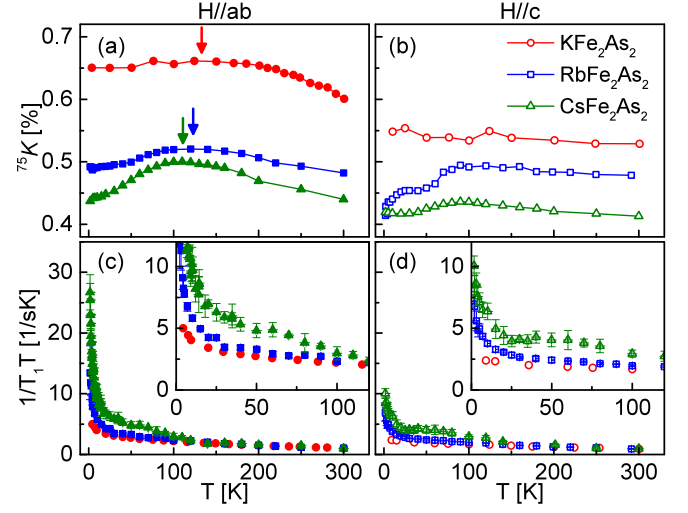


FIG. 2. NMR data of  $\text{RbFe}_2\text{As}_2$  and  $\text{CsFe}_2\text{As}_2$  reported in this work, data for  $\text{KFe}_2\text{As}_2$  are from Ref. 29. (a) and (b): Temperature dependence of the Knight shift for  $H \parallel ab$  and  $H \parallel c$ , the arrows indicate the respective maxima at  $T^*$ . (c) and (d): Temperature dependence of  $1/T_1 T$  for  $H \parallel ab$  and  $H \parallel c$ , the insets show zooms of the low-temperature data.

$\text{CsFe}_2\text{As}_2$ , thus proving the absence of a structural transition [16, 27]. The  $^{75}\text{As}$  central-line spectra of  $\text{RbFe}_2\text{As}_2$  and  $\text{CsFe}_2\text{As}_2$  for  $H \parallel ab$  and  $H \parallel c$  are shown in Fig. 1. The full width at half maximum (FWHM) is, except for  $\text{RbFe}_2\text{As}_2$  with  $H \parallel c$  [28],  $\approx 30$  kHz and only weakly temperature dependent. This is even considerably lower than the FWHM of 50 – 80 kHz reported for  $\text{KFe}_2\text{As}_2$  [9, 29], proving the very high quality of the single crystals used for our NMR study [30].

The Knight shift  $K$  probes the intrinsic uniform susceptibility. It is defined as  $K = (f_{res} - f_0)/f_0$ , where  $f_0 = \gamma_n \mu_0 H / 2\pi$  is the Larmor frequency of a bare nucleus with gyromagnetic ratio  $\gamma_n$  in a magnetic field  $H$ , and  $f_{res}$  is the observed NMR frequency, which we extract as the first moment from the recorded spectra. For  $H \parallel ab$  ( $\perp V_{ZZ}$ ), the second-order quadrupole shift  $3\nu_Q^2/16f_0$  contributes to  $f_{res}$ , but vanishes for  $H \parallel c$ . By subtracting this contribution from  $f_{res}$ , we obtain the magnetic part of the Knight shift. As shown in Fig. 2 (a) and (b), we find a similar anisotropy for all three  $\text{AFe}_2\text{As}_2$  compounds. In general, the in-plane Knight shift  $K_{ab}$  is slightly larger than the out-of-plane shift  $K_c$ , and both continuously decrease from  $\text{KFe}_2\text{As}_2$  to  $\text{CsFe}_2\text{As}_2$ . This reduction of the uniform spin susceptibility is compatible with an increase of antiferromagnetic correlations. Also,  $K_{ab}$  shows a stronger temperature dependence than  $K_c$ , yielding a broad maximum that shifts to lower temperatures and becomes increasingly pronounced.

In line with the Knight shift data, measurements of the macroscopic susceptibility of  $\text{KFe}_2\text{As}_2$  by Hardy et al. showed a maximum around 100 K, and are considered as evidence for a coherence-incoherence crossover

mechanism [14, 16]. It is suggested that localized spins dominate the susceptibility in the incoherent state at high temperatures, whereas the low-temperature coherent state is a metal with enhanced Pauli susceptibility. By defining  $T^*$  as the maximum of the temperature-dependent Knight shift for  $H \parallel ab$ , we obtain  $T^* = 133 \pm 3$  K,  $123 \pm 2$  K, and  $111 \pm 2$  K for  $\text{KFe}_2\text{As}_2$ ,  $\text{RbFe}_2\text{As}_2$ , and  $\text{CsFe}_2\text{As}_2$ , respectively.

Turning to the dynamic correlations,  $1/T_1T$  probes the fluctuating hyperfine fields at the nuclear site,  $1/T_1T \propto \sum_{q,\alpha,\beta} F_{\alpha\beta}(q) \chi''_{\alpha\beta}(q, f_{\text{res}})/f_{\text{res}}$ , with  $\alpha, \beta = \{x, y, z\}$ . Here,  $F_{\alpha\beta}$  denotes the hyperfine form factors and  $\chi''_{\alpha\beta}$  is the imaginary part of the dynamical electronic susceptibility. The low-temperature part of  $(1/T_1T)$  increases drastically along the  $\text{AFe}_2\text{As}_2$  series, as shown in Fig. 2(c) and (d). This reveals a significant increase of both in-plane and out-of-plane hyperfine-field fluctuations and, therefore, gives strong indications of an increasing dynamical susceptibility upon approaching a magnetic instability. Note that for all  $\text{AFe}_2\text{As}_2$  compounds,  $(1/T_1T)_{H\parallel c}$  is generally smaller than  $(1/T_1T)_{H\parallel ab}$ .

As was reported for  $\text{BaFe}_2\text{As}_2$  and related compounds with K, Co, or Cu doping, the anisotropy ratio of the nuclear spin-lattice relaxation rate,  $R = (1/T_1)_{H\parallel ab}/(1/T_1)_{H\parallel c}$ , allows to conclude on the  $q$  modulation of electronic spin fluctuations [29, 31]. As detailed in Refs. 29 and 32, assuming that the hyperfine field at the As site is given as the sum of the fields from the nearest-neighbor Fe spins with tetragonal symmetry (in-plane isotropy of diagonal hyperfine coupling terms, i.e.,  $A_{ab} = A_a = A_b$ ), the anisotropy ratio  $R$  is given as

$$R = \begin{cases} 0.5 + 0.5 \left( \frac{A_c S_c}{A_{ab} S_{ab}} \right)^2 & \text{no correlation} \\ 0.5 & (\pi, \pi) \\ 0.5 + \left( \frac{S_{ab}}{S_c} \right)^2 & (0, \pi) \text{ or } (\pi, 0). \end{cases} \quad (1)$$

Here,  $S_{ab}$  and  $S_c$  are the in-plane and out-of-plane components of the fluctuating Fe spins.  $R = 0.5$  corresponds to checkerboard type fluctuations with  $q = (\pi, \pi)$ . Assuming  $S_{ab} = S_c$  and taking the hyperfine coupling ratio of  $A_c/A_{ab} = 0.5$  reported for  $\text{KFe}_2\text{As}_2$  into account [29],  $R \approx 0.6$  indicates the absence of correlations, and  $R = 1.5$  results from a fully developed commensurate stripe-type [ $q = (0, \pi)$  or  $q = (\pi, 0)$ ] modulation.

Lee et al. reported two-dimensional incommensurate spin fluctuations in  $\text{KFe}_2\text{As}_2$  at  $(\pi(1 \pm 2\delta), 0)$ , with  $\delta = 0.16$ , determined by neutron scattering [33]. For all three  $\text{AFe}_2\text{As}_2$  compounds, we find, within experimental error,  $R \approx 1.2$  at 300 K, see Fig. 3. Therefore, stripe-type fluctuations, compatible with the aforementioned neutron-scattering results, where the slightly reduced value of  $R$  may stem from the incommensurability, are already well developed at room temperature. As a general trend,  $R$  increases towards low temperatures. For  $\text{KFe}_2\text{As}_2$  and

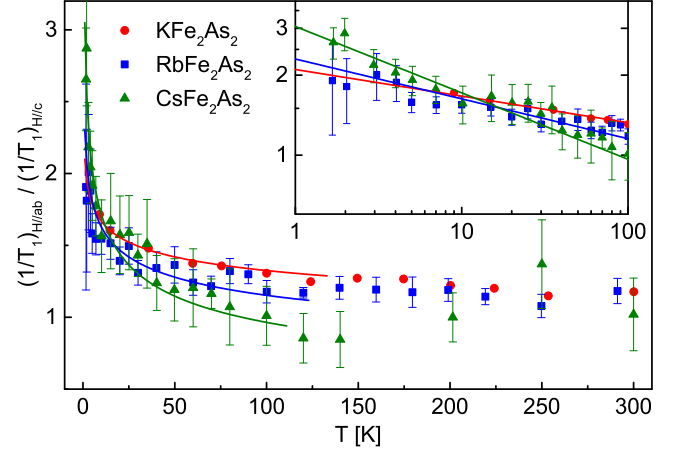


FIG. 3. Temperature-dependent anisotropy ratio  $R = (1/T_1)_{H\parallel ab}/(1/T_1)_{H\parallel c}$  for all three  $\text{AFe}_2\text{As}_2$  compounds, data for  $\text{KFe}_2\text{As}_2$  are from Ref. 29. The trend of  $R$  is, as no analytical expression is available, indicated by solid lines as a guide to the eye. The inset shows a zoom of the low-temperature data in log-log scale.

$\text{RbFe}_2\text{As}_2$ , no change of  $R$  is found at  $T^*$ , even though the temperature-dependent exponent of  $(1/T_1)$  for both applied field directions changes at  $T^*$ , as will be discussed below. For  $\text{CsFe}_2\text{As}_2$ , the  $1/T_1T$  data in Figs. 2 (c) and (d) show a hump-like structure at temperatures below about 100 K, which also seems to be reflected in a weak variation of  $R$  at intermediate temperatures. Towards lowest temperatures,  $R$  increases to  $\approx 2$  for  $\text{RbFe}_2\text{As}_2$ , and to  $\approx 3$  for  $\text{CsFe}_2\text{As}_2$ .

As was shown in Ref. 31 for Cu- and Co-doped  $\text{BaFe}_2\text{As}_2$ , in the vicinity to a quantum critical point, possibly coupled to a structural instability,  $R$  tends to increase to much larger values than 1.5. In the present case of the  $\text{AFe}_2\text{As}_2$  compounds, we have no experimental indication of a nearby structural transition. Therefore, an increase of  $R$  beyond 1.5 can only result from an increase of  $S_{ab}/S_c$ , proving the increasingly anisotropic character of correlations close to the QCP. Moreover, these findings are compatible with an electronic localization in specific Fe  $3d$  orbitals. LDA + dynamical mean field theory (DMFT) calculations have predicted an increasing localization in Fe  $3d_{z^2}$  and  $3d_{xy}$  orbitals in the  $\text{AFe}_2\text{As}_2$  series [18]. In line with these results, the study of Eilers et al. attributes a strong enhancement of effective quasiparticle masses to the hybridization of the  $3d_{xy}$  orbitals [10].

An increase of anisotropic dynamic correlations leads to an anisotropic power-law behavior with  $1/T_1 \propto T^\eta$  close to quantum criticality. Considering the given stripe-type modulation, in-plane spin fluctuations  $S_{ab}$  contribute to  $(1/T_1)_{H\parallel ab}$  via off-diagonal hyperfine coupling terms, but not to  $(1/T_1)_{H\parallel c}$  [32, 34, 35]. In consequence, mostly the power-law exponent  $\eta_{H\parallel ab}$  should evolve upon approaching a QCP driven by in-plane correlations.

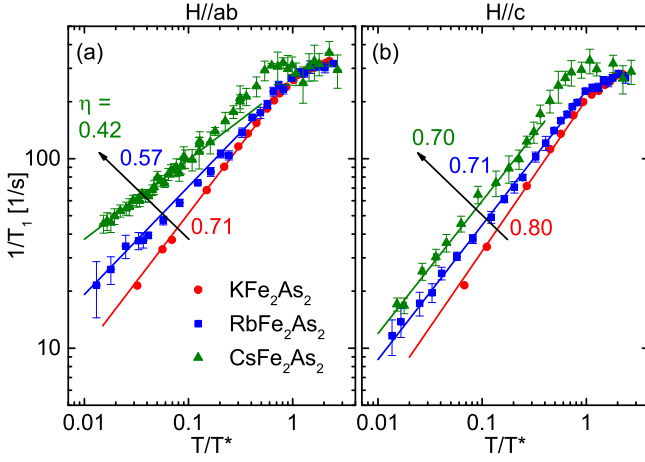


FIG. 4. The spin-lattice relaxation rate  $1/T_1$  as a function of the reduced temperature  $T/T^*$  for  $A\text{Fe}_2\text{As}_2$  ( $A = \text{K}, \text{Rb}$  and  $\text{Cs}$ ) for (a)  $H \parallel ab$  and (b)  $H \parallel c$ . The straight lines are fits with  $\propto T^\eta$  to the low-temperature part. Data for  $\text{KFe}_2\text{As}_2$  are from Ref. 29.

Along this line, we now discuss the emergent non-Fermi-liquid character of the dynamic magnetic correlations by comparing  $1/T_1$  for the  $A\text{Fe}_2\text{As}_2$  compounds for both field orientations. For better comparability, Fig. 4 shows  $1/T_1$  as a function of the reduced temperature  $T/T^*$  [36]. As shown in Fig. 4(b), all  $(1/T_1)_{H \parallel c}$  curves follow almost the same power-law relation with  $\eta_{H \parallel c}$  between 0.80 and 0.70 below  $T^*$ . For in-plane fields, however,  $\eta_{H \parallel ab}$  successively decreases from 0.71 to 0.42 below  $\approx 0.2 T/T^*$ .

In  $\text{Ba}_{0.3}\text{K}_{0.7}\text{Fe}_2\text{As}_2$ , discussed in Ref. 15,  $\eta$  is still close to unity and isotropic, as expected for a Fermi liquid. For  $\text{KFe}_2\text{As}_2$ , the reported values of  $\eta$  range from 0.75 to 0.8, indicating enhanced spin fluctuations in contrast to a standard Fermi-liquid behavior [15, 25, 37]. In the present work, we find that  $\eta$  becomes increasingly anisotropic for  $\text{RbFe}_2\text{As}_2$  and  $\text{CsFe}_2\text{As}_2$ . Moreover, the decrease of  $\eta_{H \parallel ab}$  evidences that the progressive deviation from Fermi-liquid behavior is mainly reflected by the evolution of the in-plane spin fluctuations  $S_{ab}$ . Our findings for  $\eta_{H \parallel c}$  are, therefore, in general agreement with the results reported by Wu et al., but the strong evolution of  $\eta_{H \parallel ab}$  is in sharp contrast to their statement on universality below  $T^*$  [25].

In summary, we report a detailed investigation of the anisotropic magnetic correlations in the  $A\text{Fe}_2\text{As}_2$  ( $A = \text{K}, \text{Rb}, \text{Cs}$ ) series by means of  $^{75}\text{As}$  NMR spectroscopy. The Knight-shift data reveal a successive reduction and increasing temperature dependence of the uniform spin susceptibility. The anisotropy of  $1/T_1$  allows to conclude on a stripe-type modulation of spin fluctuations up to room temperature, and an increasing low-temperature anisotropy of spin fluctuations with increasing alkali-ion radius. The power-law behavior of  $(1/T_1)_{H \parallel ab}$  gives ev-

idence that the dynamic correlations evolve towards an increasing deviation from Fermi-liquid behavior, reflected mostly by the in-plane spin fluctuations. All these findings clearly indicate that the electronic correlations in the  $A\text{Fe}_2\text{As}_2$  series evolve towards a quantum critical point at the transition to a yet unobserved, magnetically ordered phase.

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